High-resolution mapping of nitrogen oxides emissions in US large cities from TROPOMI retrievals of tropospheric nitrogen dioxide columns

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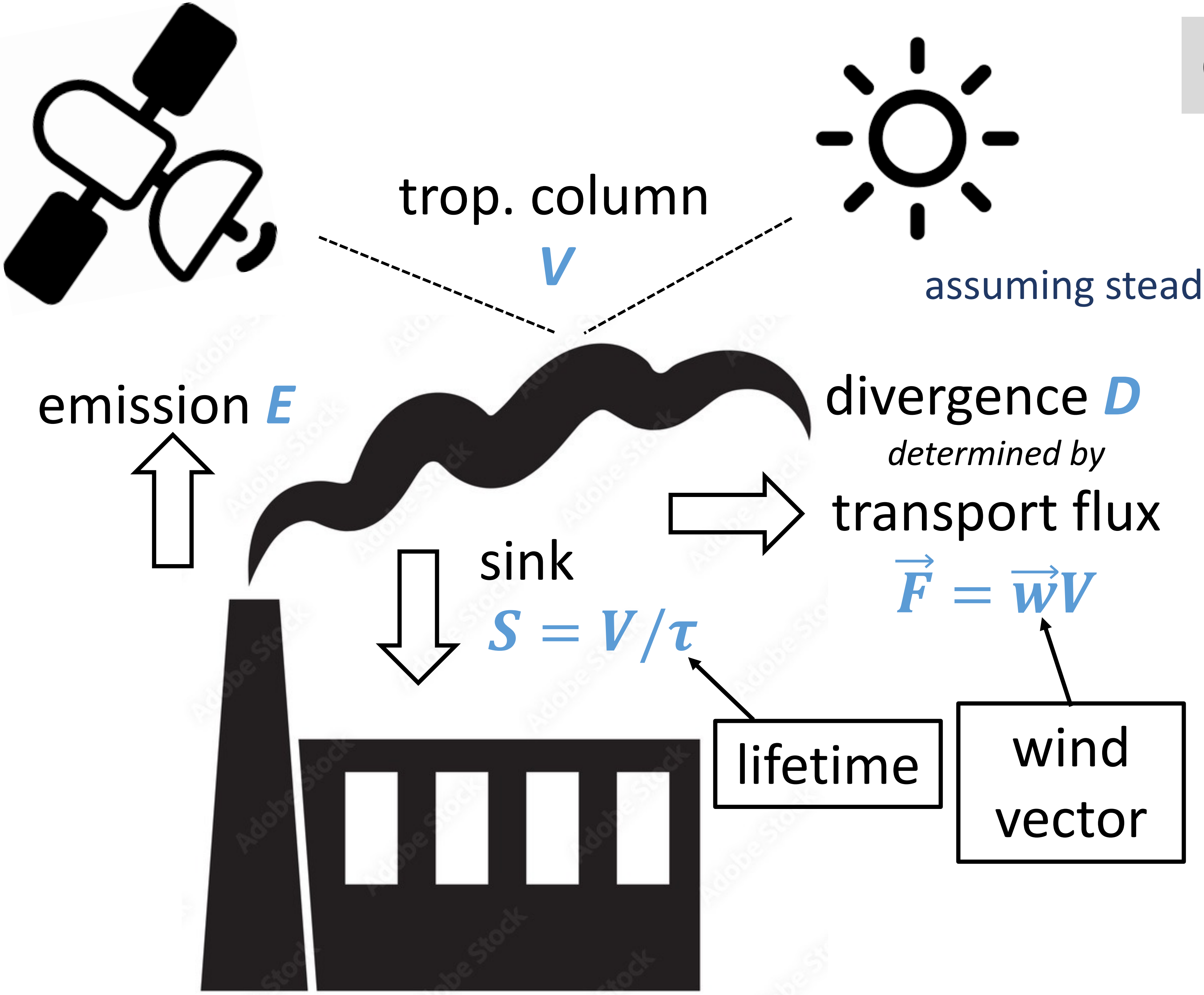
continuity equation

$$\frac{\partial V}{\partial t} = E - S - D$$

assuming steady state: $0 = E - S - D$

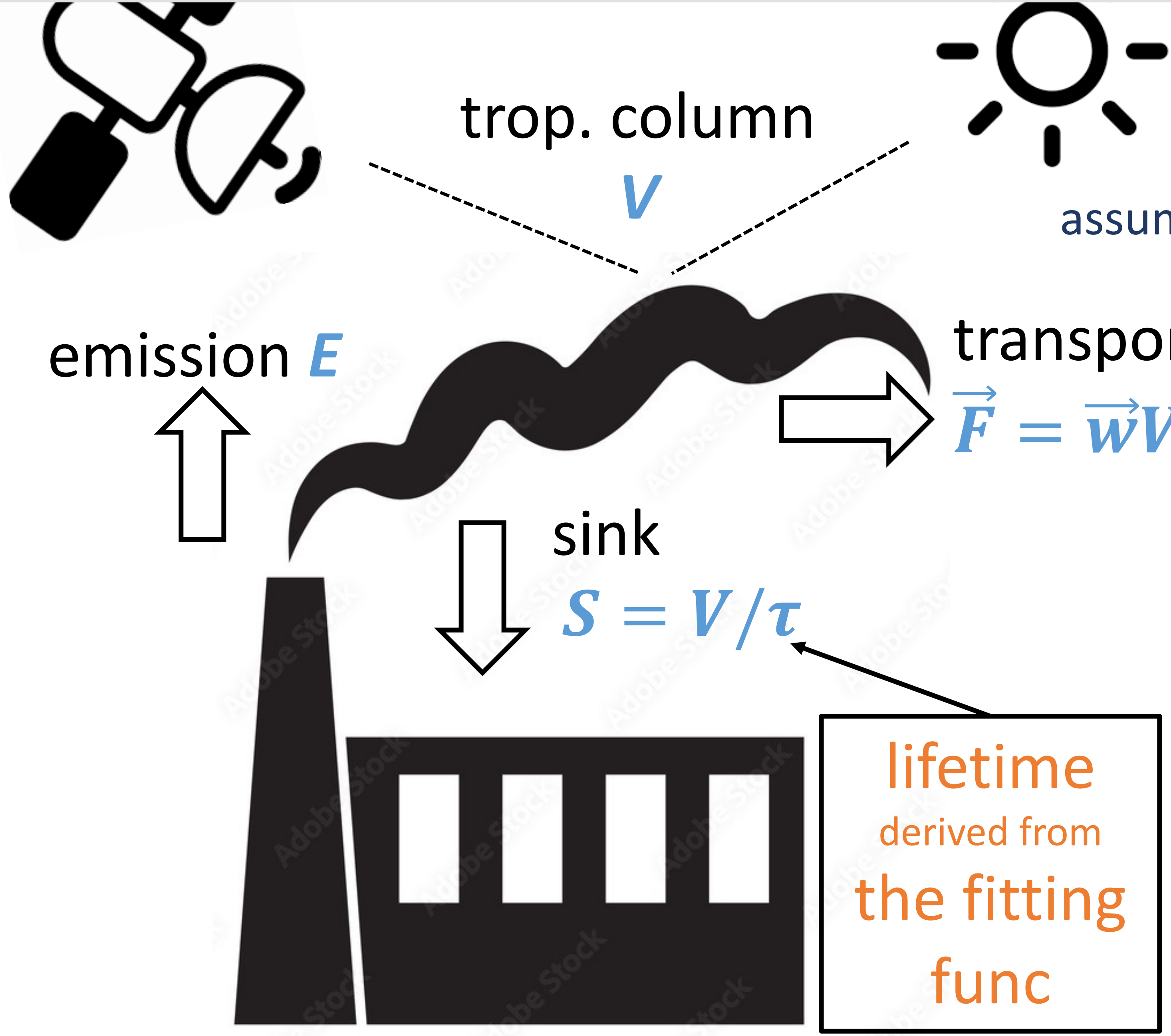
$$\begin{aligned} E &= D + S = \nabla \cdot \vec{F} + S \\ &= \nabla \cdot \vec{w}V + V/\tau \end{aligned}$$

$$\begin{aligned} mean(E) &= mean(D) + mean(S) \\ &= \nabla \cdot mean(\vec{F}) + mean(S) \\ &= \nabla \cdot mean(\vec{w})mean(V) + mean(V)/\tau \end{aligned}$$



Beirle, S., Borger, C., Dörner, S., Li, A., Hu, Z., Liu, F., Wang, Y., and Wagner, T.: Pinpointing nitrogen oxide emissions from space, Science Advances, 5, eaax9800, https://doi.org/10.1126/sciadv.aax9800, 2019.

Continuity equation of NO_2 columns



$$\frac{\partial V}{\partial t} = E - S - D$$

assuming steady state: $0 = E - S - D$

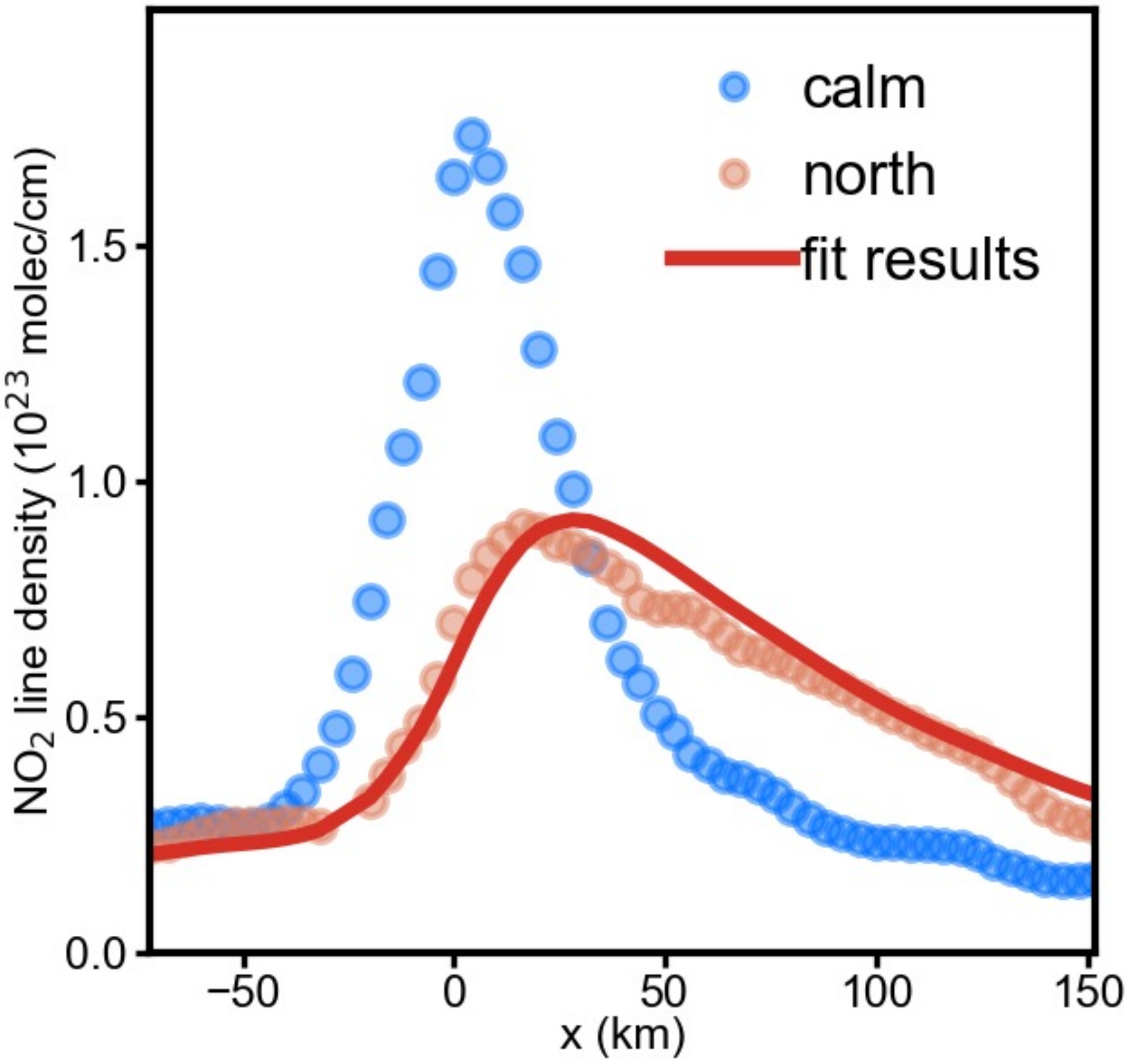
$$E = D + S = \nabla \cdot \vec{F} + S$$
$$= \nabla \cdot \vec{w}V + V/\tau$$

anthropogenic

$$mean(E_{ant}) = mean(D_{ant}) + mean(S_{ant})$$
$$= \nabla \cdot mean(\vec{w}) mean(V - b) + mean(V - b)/\tau$$

b: lowest 1th percentile of tropospheric NO_2 vertical column densities under calm wind condition over the study area (150km*150 km)

Fitting function for lifetime



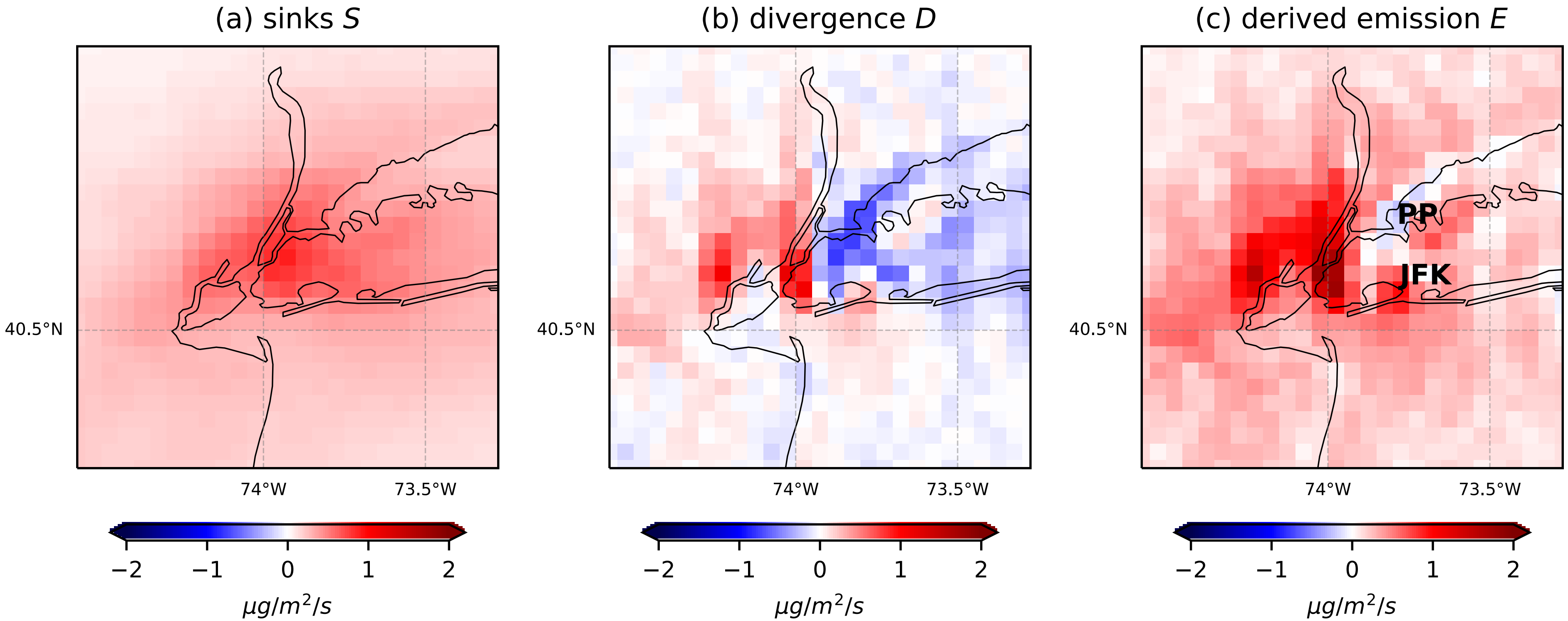
Perform a **nonlinear least-squares fit** of *f(x)* to the observed line densities under windy conditions $LD_{windy}(x)$ with τ as the fitting parameter

- Assuming each grid cell releases a constant NO_x emission rate *E(x)*
- Wind is blowing continuously in a direction x with a speed *w* $\frac{E(x)}{ratio \times w}$
- NO_x reactions follow exponential decay $e^{-\frac{x}{w \times \tau}}$

$$f(x) = \frac{E(x)}{ratio \times w} * e^{-\frac{x}{w \times \tau}} + b'$$
$$= \frac{[LD_{calm}(x) - b]}{w \times \tau} * e^{-\frac{x}{w \times \tau}} + b'$$

Liu, F., Tao, Z., Beirle, S., Joiner, J., Yoshida, Y., Smith, S. J., Knowland, K. E., and Wagner, T.: A new method for inferring city emissions and lifetimes of nitrogen oxides from high-resolution nitrogen dioxide observations: a model study, Atmos. Chem. Phys., 22, 1333–1349, https://doi.org/10.5194/acp-22-1333-2022, 2022.

Case study: New York



$$mean(E_{ant}) = \nabla \cdot mean(\vec{w})mean(V - b) + mean(V - b)/\tau$$

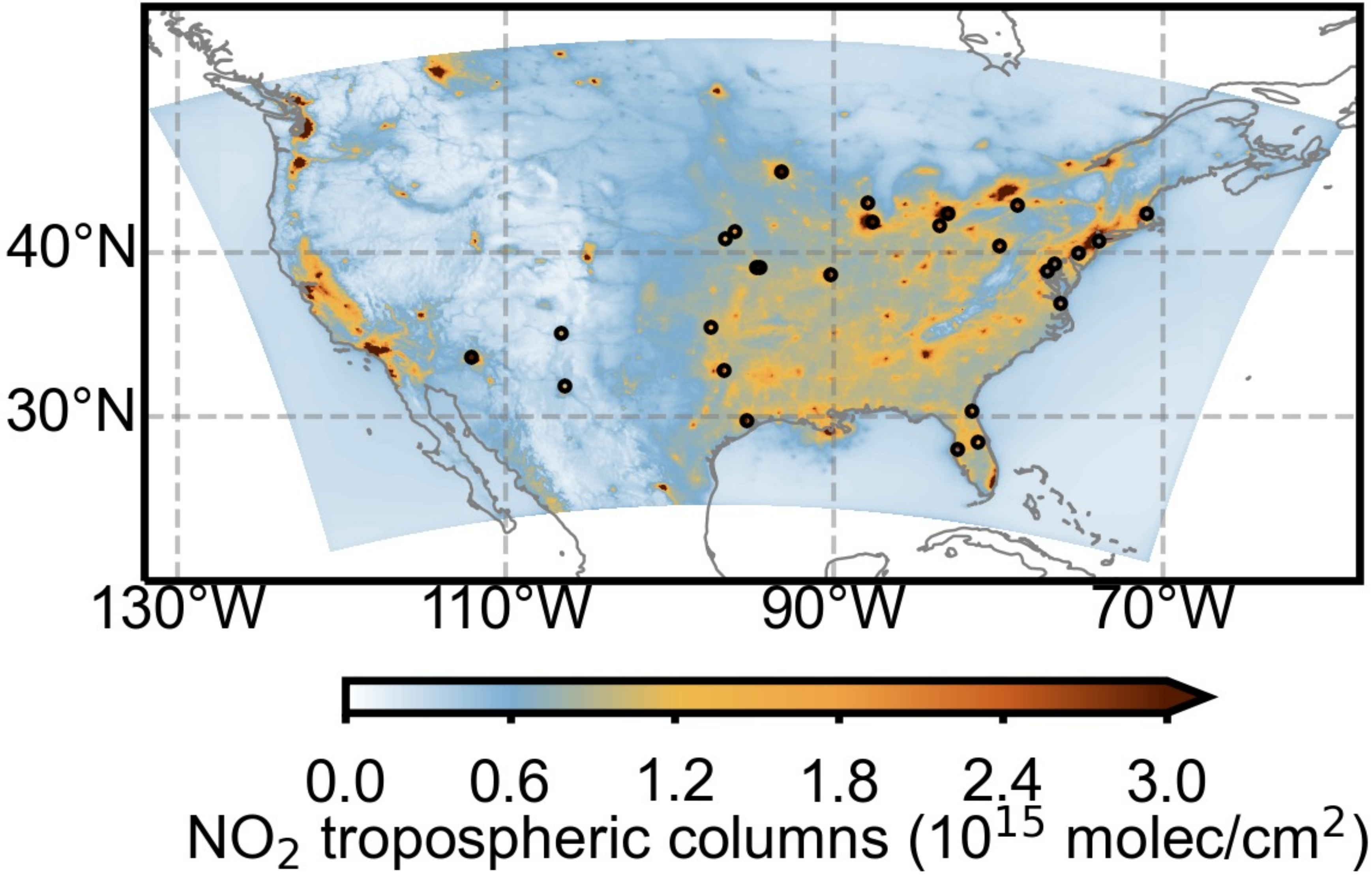
trop. column ***V***: TROPOMI GSFC NO_2 retrieval (May - September, 2019); qa > 0.75

wind field $\vec{w}$: GEOS FP-IT reanalysis wind; Interpolated to orbit timestamp; Averaged at 1000 m above ground

Fitted lifetime τ: fit based on TROPOMI GSFC NO_2 retrieval (May - September, 2018-2021)

$[NO_x]/[NO_2] = 1.32$

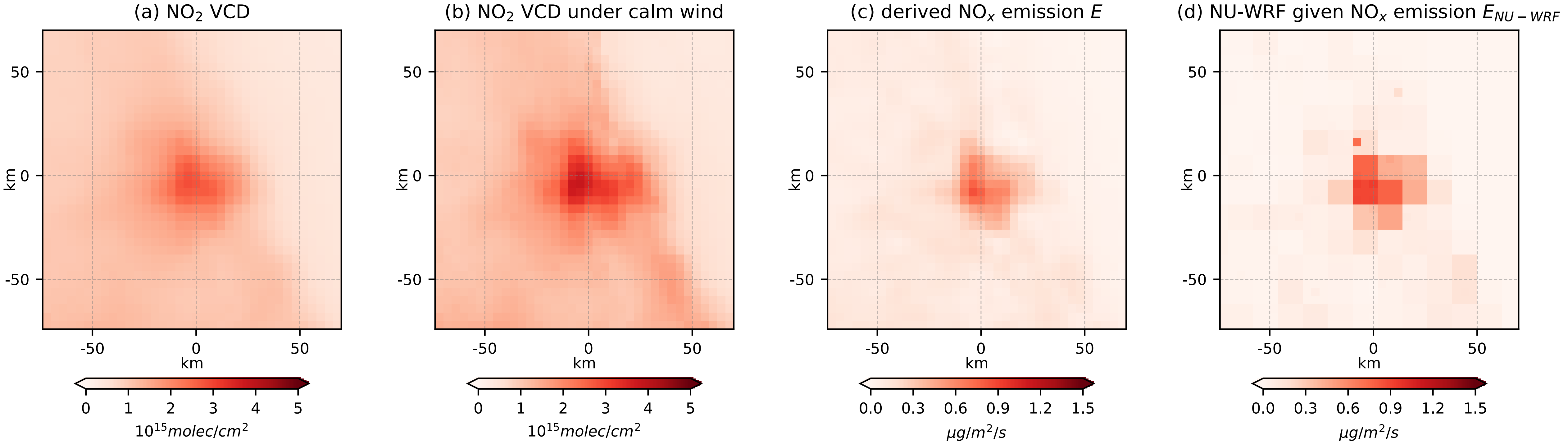
Validation using model data



- Model: NU-WRF generates synthetic satellite observations
- Time: May to September, 2016
- Spatial resolution: 4 km (comparable to TROPOMI and TEMPO)
- Select all US major cities with population > 200,000

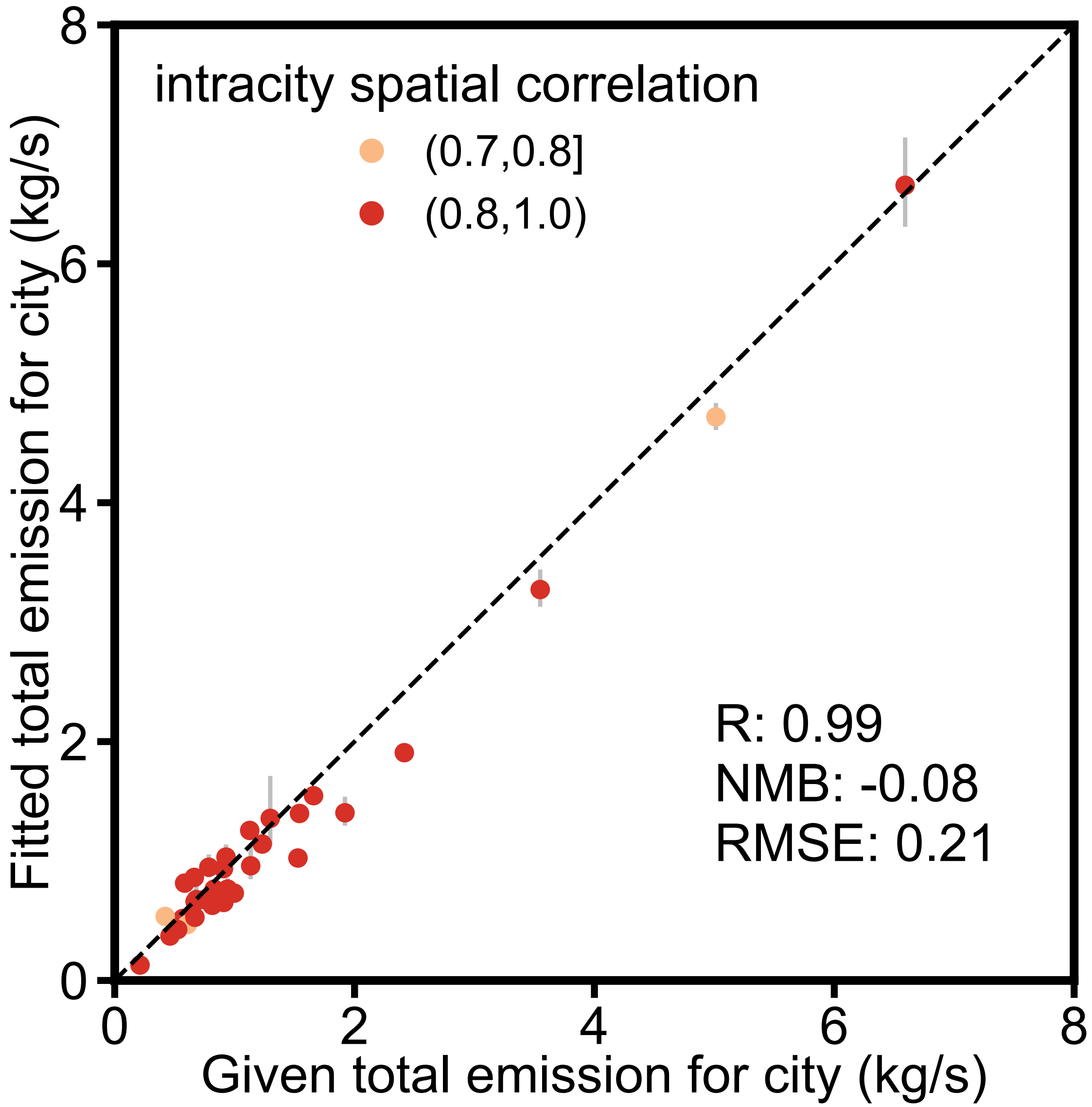
Improved intracity spatial correlation

Jacksonville



R: 0.75 0.80 0.92

Validation results



- Fit of lifetime works for 41 out of 70 US large cities with R > 0.9, root-mean-square deviation (RMSD) < 10%, fitted error of τ < 10%
- Only keep background (i.e., 1 percentile of calm-wind NO_2 in the 300 * 300 km domain) / averaged (i.e., average calm-wind NO_2 over the urban area) < 50%, which left 33 cities
- urban areas used for calculation emission in scatter plot:

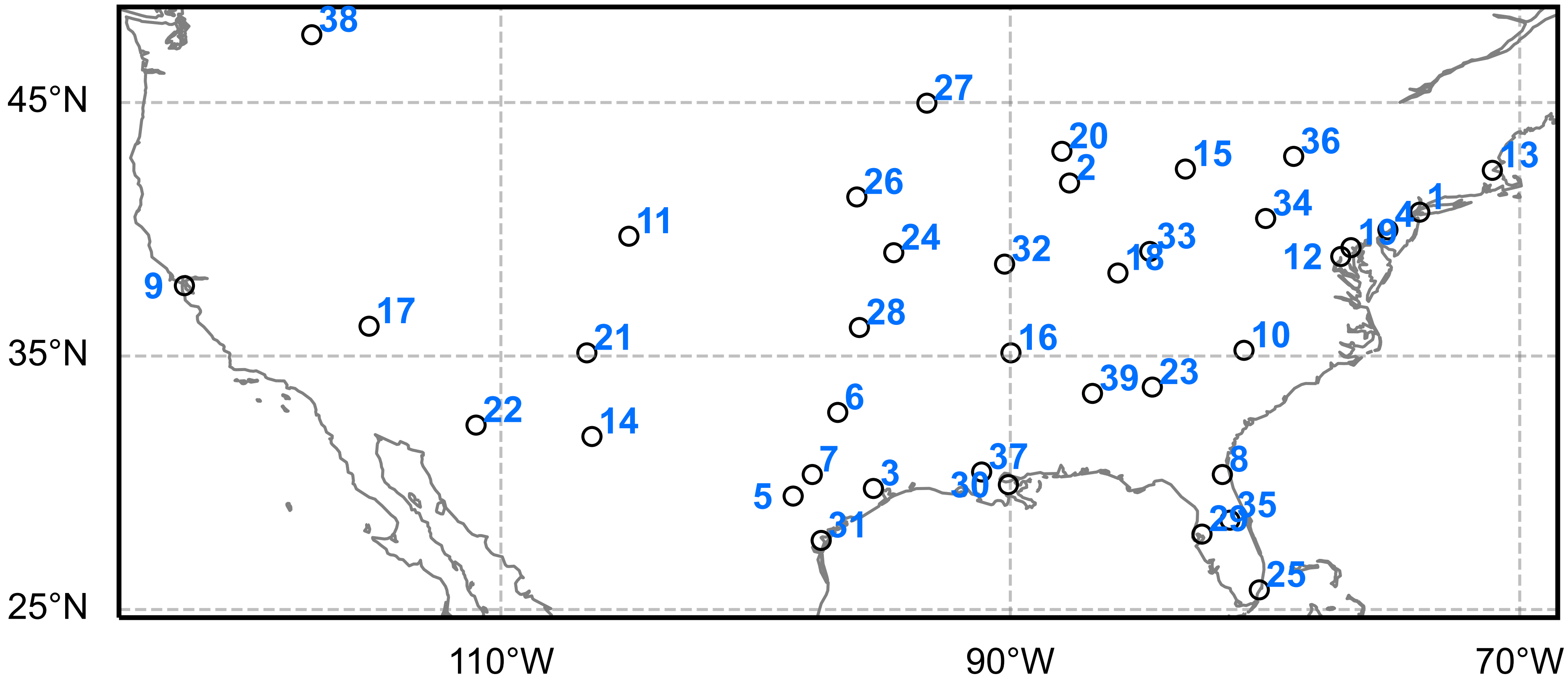
New York, Chicago, Los Angeles and Houston: 100*100 km
Other cities: 70*70 km

- the differences between fitted and given emissions: -8 ± 18%

For 150*150 km^2 domain around city center:

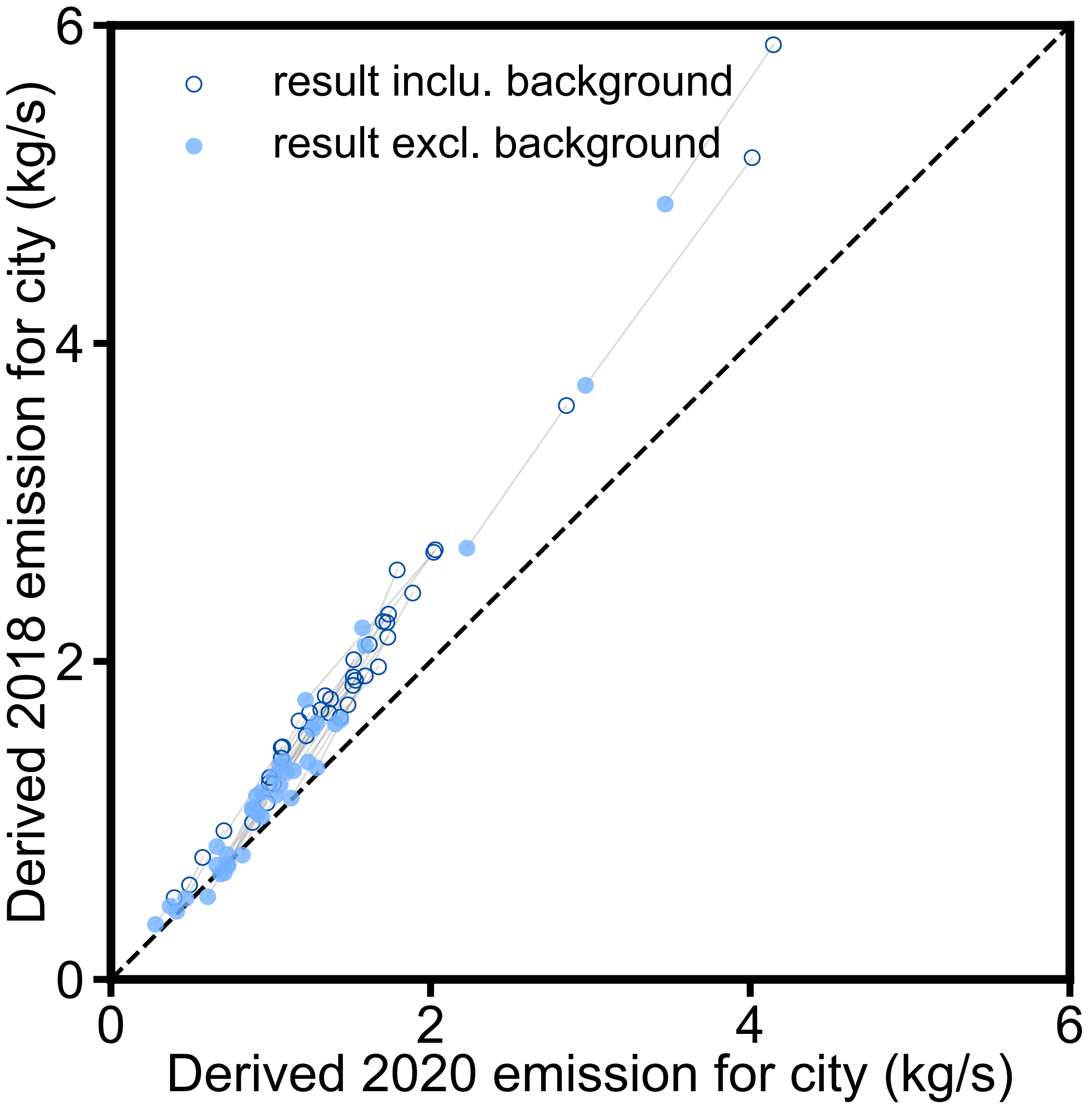
- correlation coefficient of given emissions vs fitted emissions: 0.88 ± 0.06
- correlation coefficient of given emissions vs VCD: 0.78 ± 0.09
- correlation coefficient of given emissions vs calm-wind VCD: 0.80 ± 0.08

Spatial distribution of investigated cities



- Fit of lifetime works for 53 out of 70 US large cities: R > 0.9, root-mean-square deviation (RMSD) < 10%, fitted error of τ < 10%
- Discard cities with background *b* / averaged NO_2 > 50%, which left 39 cities

Annual variation of emissions

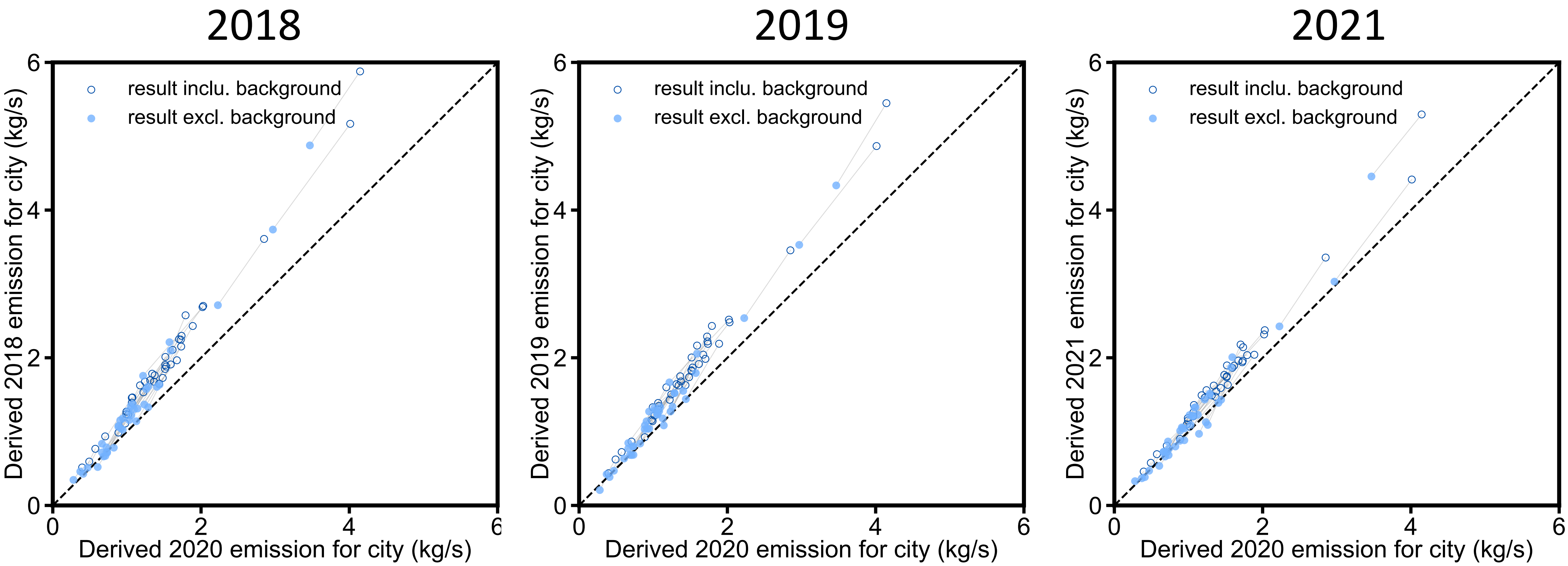


result inclu. background: $mean(E)$
$= \nabla \cdot mean(\vec{w})mean(V) + mean(V)/\tau$

result excl. background: $mean(E_{ant}) =$
$\nabla \cdot mean(\vec{w})mean(V-b) + mean(V-b)/\tau$

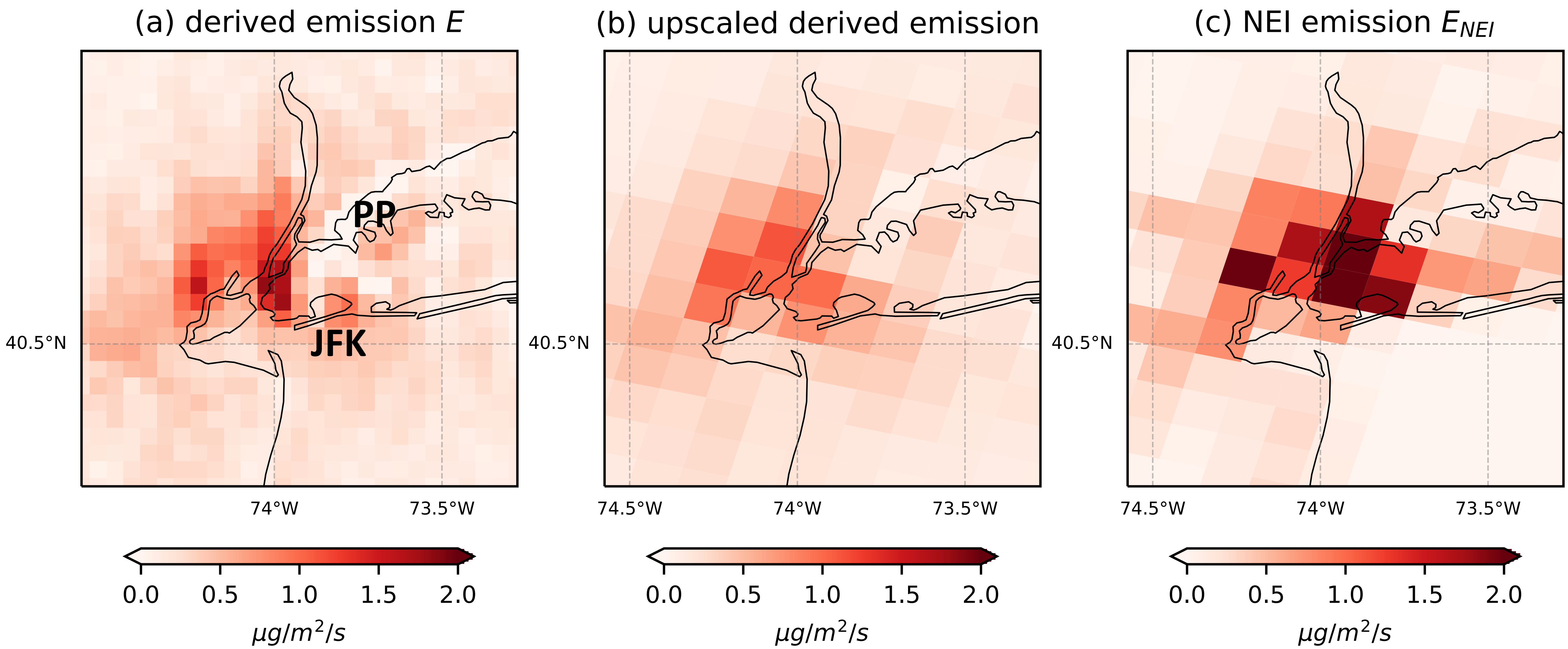
lifetime τ : multiple-year averaged value based on 2018-2021 TROPOMI GSFC NO_2 retrieval (May - September)

Annual variation of emissions



lifetime τ: multiple-year averaged value based on 2018-2021 TROPOMI GSFC NO_2 retrieval (May - September)

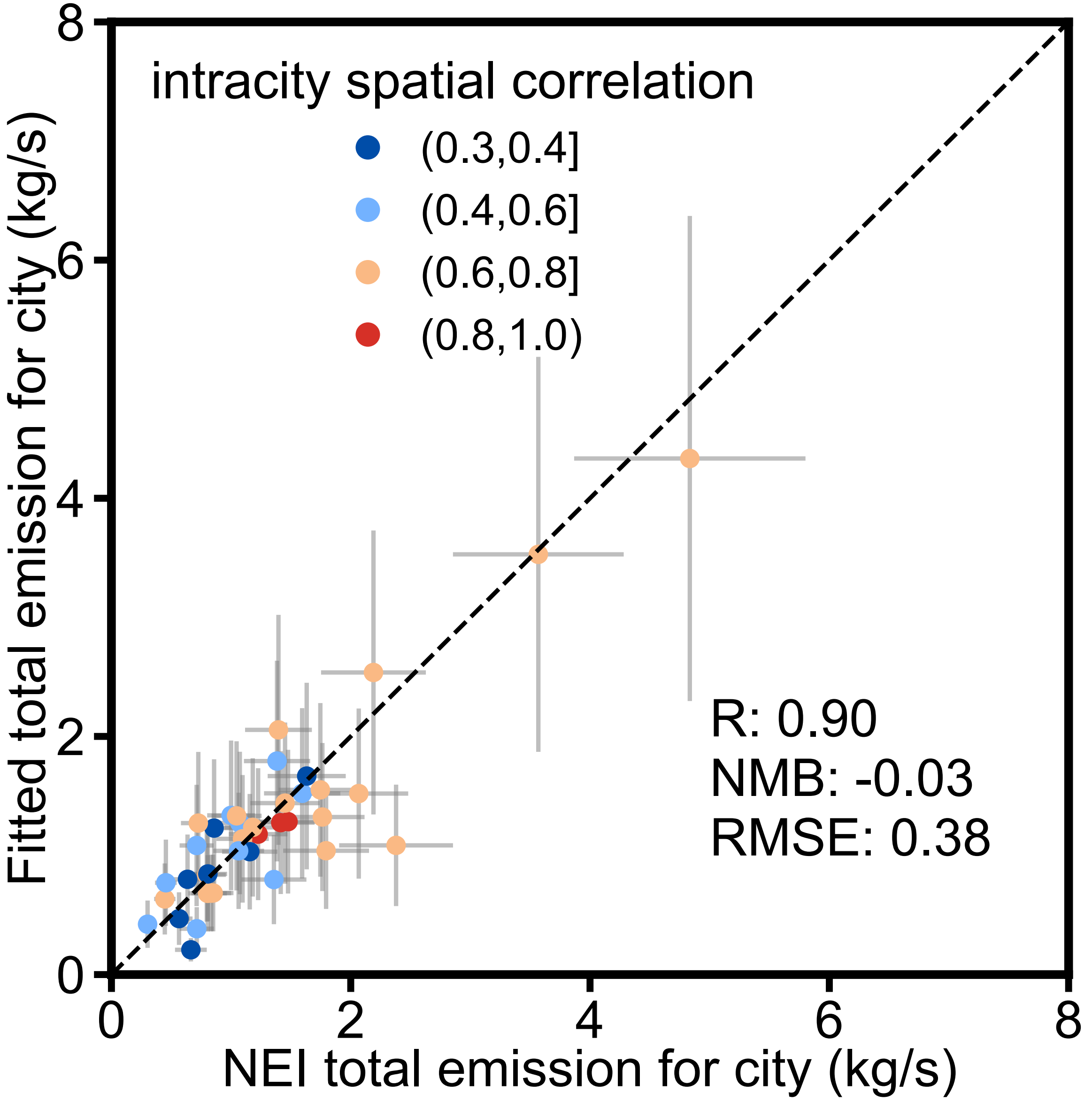
Compare with NEI



NEI E_{NEI}: year 2019; spatial resolution of 12 km

TROPOMI-based E: year 2019; spatial resolution of 0.05 degree

Compare with NEI



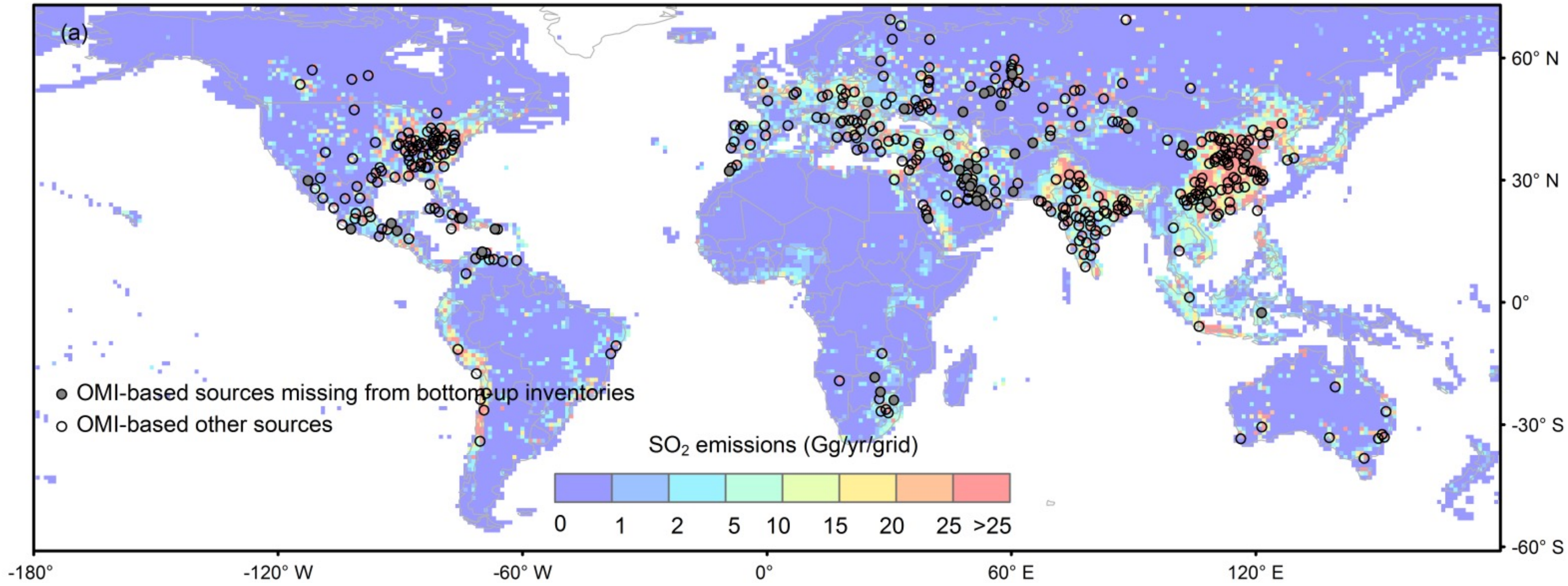
- Urban areas used to calculate total emission in scatter plot:

New York, Chicago, Los Angeles and Houston: 100*100 km
other cities: 70*70 km

- the differences between fitted and given emissions:
  3 ± 32%

- correlation coefficient of given emissions vs fitted emissions:
  0.59 ± 0.15

fusion emission inventory



$$$ funded by ACMAP program

- Combine **Sat**ellite-derived SO_2 emissions for large point sources with a bottom-up inventory **CEDS** derived from reported fossil fuel combustion for smaller sources, to construct a new inventory **CEDS-SatEm**
- Data has been released: https://zenodo.org/record/6964915#.YzOmhOxq30o
- Spatial resolution of 0.1/0.5 degree

Propose:
a fusion emission inventory reconciling satellite-derived NO_x emissions with CEDS to provide long-term, global anthropogenic **spatiotemporally-resolved** emissions of NO_x and co-emitted air pollutants updated to the most recent year to support fine-resolution simulations of tropospheric composition

Liu, F., Choi, S., Li, C., Fioletov, V. E., McLinden, C. A., Joiner, J., Krotkov, N. A., Bian, H., Janssens-Maenhout, G., Darmenov, A. S., and da Silva, A. M.: A new global anthropogenic SO_2 emission inventory for the last decade: a mosaic of satellite-derived and bottom-up emissions, Atmos. Chem. Phys., 18, 16571-16586, https://doi.org/10.5194/acp-18-16571-2018, 2018.

Take home message

We develop a new dataset of gridded NO_x emissions for major US cities, which is:

- High spatial resolution of 0.05 degree
- Chemical transport model-independent
- Annually updated
- Extended globally